



(19)

Europäisches Patentamt

European Patent Office

Office européen des brevets



(11)

EP 1 304 044 A2

(12)

## EUROPEAN PATENT APPLICATION

(43) Date of publication:  
23.04.2003 Bulletin 2003/17

(51) Int Cl.7: A23L 1/22, A23P 1/08,  
A23P 1/12, A23G 3/00,  
C11D 3/50

(21) Application number: 02257279.6

(22) Date of filing: 21.10.2002

(84) Designated Contracting States:  
AT BE BG CH CY CZ DE DK EE ES FI FR GB GR  
IE IT LI LU MC NL PT SE SK TR  
Designated Extension States:  
AL LT LV MK RO SI

(30) Priority: 22.10.2001 US 54239

(71) Applicant: INTERNATIONAL FLAVORS &  
FRAGRANCES INC.  
New York New York 10019 (US)

(72) Inventors:  
• Lou, Wen Chin  
Morris, New Jersey 07950 (US)  
• Popplewell, Lewis Michael  
Monmouth, New Jersey 07751 (US)

(74) Representative: Mercer, Christopher Paul et al  
Carpmaels & Ransford  
43, Bloomsbury Square  
London WC1A 2RA (GB)

### (54) Hydroxypropyl cellulose encapsulation material

(57) Hydroxypropyl cellulose is employed to encapsulate flavor or fragrance materials that could be employed in diverse applications such as food products and laundry applications.

EP 1 304 044 A2

**Description****Field of the Invention**

5 [0001] The present invention relates to an encapsulation composition in which the encapsulate, preferably a flavor or fragrance material, is encapsulated in a matrix containing hydroxypropyl cellulose.

**Background of the Invention**

10 [0002] As described in U.S. Patent No. 6,187,351 ('351 patent), the encapsulation of various materials such as medications, pesticides, flavors and fragrances is an ongoing area of research. The '351 patent, hereby incorporated by reference in its entirety, provides an overview of the art used to incorporate various materials. The '351 patent adds to this body of work by disclosing the use of various food polymers, maltodextrin, and a saccharide, disaccharide or corn syrup material. The food polymers disclosed in the patent are set forth in column 10, lines 29-37.

15 [0003] Despite the disclosure of the above patent and the numerous patents cited therein, there remains a continuing need to provide materials in encapsulated form that are stable at elevated temperatures and/or dissolve slowly in aqueous environments, particularly for volatile materials, such as flavors and fragrances.

**Summary of the Invention**

20 [0004] One embodiment of the invention is a composition comprising:

a flavor or fragrance encapsulate encapsulated in a matrix, the matrix which may be amorphous or partially crystalline, the matrix comprising:

25 from about 60 to about 99.5 weight percent of materials selected from the group consisting of sugars, maltodextrin having 5 to 20 dextrose equivalent (DE), fats, silicone dioxide, polyhydric alcohols, corn syrup solids, starches, modified starches, emulsifiers and food acids;

30 from about 0.5 to about 20 weight percent, preferably from about 2 to about 10 weight percent of hydroxypropyl cellulose; and

from about 0.1 to about 20 weight percent of a flavor or fragrance material.

[0005] In a second embodiment of the invention the above composition is prepared by a process that comprises the following steps of mixing the components together, optionally with a liquid plasticizer, and providing a heating source such as an extruder to obtain a melted matrix and then creating the desired particle size by forming, cooling, and sizing operations.

[0006] These and other embodiments of the present invention will become apparent by reading the detailed description and accompanying examples.

**40 Detailed Description of the Invention**

[0007] As used herein, flavor is understood to include spice oleoresins derived from allspice, basil, capsicum, cinnamon, cloves, cumin, dill, garlic, marjoram, nutmeg, paprika, black pepper, rosemary, and turmeric, essential oils, anise oil, caraway oil, clove oil, eucalyptus oil, fennel oil, garlic oil, ginger oil, peppermint oil, onion oil, pepper oil, rosemary oil, spearmint oil, citrus oil, orange oil, lemon oil, bitter orange oil, tangerine oil, alliaceous flavors, garlic, leek, chive, and onion, botanical extracts, arnica flower extract, chamomile flower extract, hops extract, marigold extract, botanical flavor extracts, blackberry, chicory root, cocoa, coffee, kola, licorice root, rose hips, sarsaparilla root, sassafras bark, tamarind and vanilla extracts, protein hydrolysates, hydrolyzed vegetable proteins, meat protein hydrolyzes, milk protein hydrolyzates and compounded flavors both natural and artificial including those disclosed in S. Heath, *Source Book of Flavors*, Avi Publishing Co., Westport Connecticut, 1981, pages 149-277.

[0008] Many types of fragrances can be employed in the present invention, the only limitation being the compatibility with the other components being employed. Suitable fragrances include but are not limited to fruits such as almond, apple, cherry, grape, pear, pineapple, orange, strawberry, raspberry; musk, flower scents such as lavender-like, rose-like, iris-like, and carnation-like. Other pleasant scents include herbal and woodland scents derived from pine, spruce and other forest smells. Fragrances may also be derived from various oils, such as essential oils, or from plant materials such as peppermint, spearmint and the like.

[0009] A list of suitable fragrances is provided in U.S. Patent No. 4,534,891, the contents of which are incorporated by reference as if set forth in its entirety. Another source of suitable fragrances is found in Perfumes, Cosmetics and

Soaps, Second Edition, edited by W. A. Poucher, 1959. Among the fragrances provided in this treatise are acacia, cassie, chypre, cyclamen, fern, gardenia, hawthorn, heliotrope, honeysuckle, hyacinth, jasmine, lilac, lily, magnolia, mimosa, narcissus, freshly-cut hay, orange blossom, orchid, reseda, sweet pea, trefle, tuberose, vanilla, violet, wall-flower, and the like.

- 5 [0010] When the encapsulate is lipophilic, the encapsulate is dispersed in the amorphous matrix of the final product usually with the aid of an emulsifier added to the lipophilic phase or in the matrix mixture. In contrast, when the matrix is hydrophilic, or water soluble, the final product contains the encapsulate as a dissolved solute and/or as a dispersed encapsulate. It is preferred that the elements of the invention are thoroughly heated and most preferably melted in order to form the matrix. However, the present invention may still be carried out if all of the elements are not melted.
- 10 [0011] The final structure of the matrix, i.e., whether the matrix is crystalline or amorphous, will depend on the materials contained in the matrix as well as how the elements were admixed and melted.
- 15 [0012] The amount of the encapsulate provided in the matrix will depend on various factors including in part on the nature of the matrix, and the end use of the mixture. Typically the final composition will contain from about 2.5 to about 15 weight percent of encapsulate, preferably from about 4 to about 12 and most preferably from about 5 to about 10 weight percent of the encapsulate.
- 20 [0013] The matrix is comprised of one or more of the following materials: sugars, maltodextrin having 5 to 20 dextrose equivalent (DE), fats, silicone dioxide, polyhydric alcohols, corn syrup solids, starches, modified starches, emulsifiers and food acids.
- 25 [0014] The level of maltodextrin used in the matrix, comprises from about 25 to about 98 weight percent, preferably from about 35 to about 75 weight percent, the maltodextrin preferably having a DE of from about 5 to about 20. Preferably the maltodextrin has a DE of from 10 to about 15. As is generally appreciated by those with skill in the art, maltodextrin is commercially supplied with a water content of from about 4 to about 7 weight percent. Additional water may also be added with the addition of various plasticizers or with any of the other components used in the present invention.
- 30 [0015] Suitable food acids useful in the invention include citric acid, tartaric acid, malic acid and the like. The level of food acid can vary greatly from 1 to about 80 weight percent, preferably from 5 to about 60 and most preferably from 10 to about 40 weight percent. The encapsulation technology of the present invention provides the food acid from dispersing in the matrix. As a result, the food acids provide an improved tartness to the food products which contain the food acid.
- 35 [0016] In addition to the maltodextrin, other materials can be used in the matrix such as starches and modified starches. In a preferred embodiment, the level of starches is from about 5 to about 75 weight percent, preferably from 10 to about 60 and most preferably from about 15 to about 50 weight percent.
- 40 [0017] Mono and di-saccharides suitable for use in the invention include glucose, fructose, lactose, galactose, ribose, xylose, sucrose, maltose, polyols such as glycerin and propylene glycol, as well as corn syrups may also be employed. The level of the mono and di saccharides range from about 1 to about 60, preferably from about 4 to about 45 and most preferably from about 10 to about 30 weight percent of the composition.
- 45 [0018] Other ingredients such as food emulsifiers including monoglycerides of fatty acids, distilled succinylated monoglycerides of fatty acids, sorbitan fatty acid esters; distilled acetylated monoglycerides of fatty acids, monoglycerides of fatty acids and fats and oils from food lipid sources can be added to the matrix. The emulsifiers are typically added in amounts at levels of from about 0.25 to about 2.5 weight percent of the encapsulations.
- 50 [0019] In addition to the above-recited materials, other materials conventionally used in the art can be employed in the matrix of the invention. Suitable materials include but are not limited to colorings, sweeteners, diluents, fillers, preservatives, anti-oxidants, stabilizers, lubricants and the like.
- [0020] In the present invention, hydroxypropyl cellulose (HPC) is employed. The HPC useful in the invention having a viscosity of from about 3 to about 100,000 centipoise, preferably from about 4,000 to about 15,000 centipoise. The viscosity is measured at 20°C and 2 weight percent solution. The level of HPC is from about 0.5 to about 20 weight percent, preferably from about 2 to about 10 weight percent and most preferably from about 3 to about 5 weight percent.
- 55 [0021] The manufacture of the encapsulation materials of the present invention can be thought of comprising three process steps of: forming, cooling and sizing.
- [0022] The forming process involves the admixture of the components and heating of the mixture. It is preferable to have the contents melted in order to have a uniform mixture and consistency of product. The flavor or fragrance may then be added to the mixture either before or during the heating process and then admixed to provide consistency.
- [0023] The mixture containing the flavor or fragrance is then cooled using any appropriate means of convective,

conductive, or evaporative heat removal. The cooled material is then sized using conventional processes such as chopping or grinding of the material. Size classification can be done to remove fines or oversized particles. In a preferred embodiment, the sizing of the particles is done by an extrusion process in which a strip or rod-like material is formed followed by a cutting or chopping process.

5 [0024] The present invention provides a flavor or fragrance system that can withstand the high-temperature/high-moisture environments encountered in certain food-processing operations yet still allow release of the flavor/fragrance active when the product is consumed or used. The encapsulation method of the present invention allows the encapsulated flavor or fragrance to withstand vigorous processing conditions without dissolving into the bulk system or volatilizing. The robust nature of the encapsulated materials are most readily obtained when the water levels in the product composition are minimized, less than 10 weight percent of the total composition, more preferably less than 5 and most preferably less than 3 weight percent water in the total composition. Depending on the level of the water contained in the composition and the formulation of the composition, it is possible to have the encapsulated flavor or fragrance material remain encapsulated at temperatures greater than about 130, preferably above 140 and most preferably 150°C. The higher temperatures allow the materials to be processed at higher temperatures without volatilizing the flavor or fragrance materials.

10 [0025] The matrix system described in the invention can be employed in various products such as hard, gummy and chewy candies representing a range of temperature/moisture conditions. Other applications included snack foods, cereals, baked goods, pasta snacks, dairy foods and gums in which the flavor could be encapsulated in the matrix and not released until the matrix is compromised. An additional advantage of the invention is the enhanced visual appeal 15 in the finished product since the flavor particle can remain intact if the matrix and product processing conditions are properly selected.

20 [0026] In addition to food products, the present invention has utility in fragrance applications such as laundry detergent powders and laundry detergent tablets. Due to the slower solubility of the particles in washing conditions, the fragrance would be delivered later in the wash cycle providing delayed release of fragrance and ultimately improved 25 deposition and substantivity. Additional possible uses of the slow-dissolving product include granular dishwashing detergents, antiperspirants, and deodorants.

25 [0027] In the method of the present invention, the flavor or fragrance material is encapsulated with a matrix of HPC and sugars or maltodextrins, or other carrier materials. The flavor or fragrance may be in the form of a solid, liquid or paste. Preferably, the active agent is in the form of a liquid or solid, most preferably a liquid. The encapsulation may 30 be carried out by any conventional method, including spray drying, melt extrusion, and freeze drying. These methods are set forth in greater detail in U.S. Patent No. 6,245,366, the contents are incorporated by reference as if set forth in its entirety. The preferred method is to extrude the material and then cool and size the material to achieve the desired granulation.

35 [0028] The chemical and physical properties of the encapsulated active ingredient will depend on the matrix and method used for encapsulation as well as the identity of the active agent itself. For example, encapsulated active ingredients prepared by spray drying will have a particle size of from about 5 to about 200 microns, and a solid density of from about 1 to about 1.4 grams per cubic centimeters. Other features of the invention will become apparent in the course of the following descriptions of exemplary embodiments that are given for illustration of the invention and are not intended to be limited thereof.

#### Example 1

40 [0029] A formula containing carriers such as sugars, maltodextrins, fats and a HPC, some containing an encapsulated flavor or fragrance and some not containing either flavor or fragrance, was subjected to temperatures of from 100 to 190°C at low moisture conditions for a short period of time, about 2 minutes on average. The heat treated mass was cooled and sized to create solid flavoring particles or granules. The granules/particles will not dissolve or substantially deform in hard candy applications that reach approximately 290°F at 90-98% total solids. Additionally in other confectionery systems processed at 50-80% solids and 140-220°F (gummy candies, chewy candies) the products did not dissolve or dissolved at a very slow rate, thus allowing the products to survive the manufacturing process largely intact. 45 It is hypothesized that the slower solubility will improve the flavor/active delivery in higher moisture food systems as well as delivery of fragrances in non-food applications.

#### Example 2

50 [0030] Three versions of a cherry flavor were made via melt extrusion. Each flavor contained from about 3 to about 5 weight percent of flavor, about 2 weight percent lake color blend (blend of Red #40 and Blue #1 lakes) and 5 weight % acid blend (blend of citric and malic acid) as actives. The balance of the formulation contained a common blend of maltodextrin GPC 15 DE, M150 sucrose, cottonseed oil and silicone dioxide. The level of HPC was allowed to vary.

5 [0031] The HPC levels were varied as follows:

Sample A	No HPC
Sample B	2 weight percent HPC (Hercules KLUCEL® GF)
Sample C	3 weight percent HPC (Hercules KLUCEL® GF).

10 [0032] Each formulation was extruded under similar conditions, cooled and then sized to yield -10/+14 granules.

[0033] The granules were added to the following hard candy formulation and tested for stability:

Sugar, fine granule weight %	56
Corn syrup 43 BE	27
Tap water	17
Total	100

15 [0034] The sugar, corn syrup and water were weighed into a 400 milliliter beaker and stirred together. The ingredients were heated in a 1000 watt microwave for approximately 105 seconds, until the temperature of the mixture reaches approximately 300°F. The beaker was removed from the microwave and was briefly stirred. The extruded flavor was added and blended until uniform. The melt was then deposited into moulds and allowed to cool. Once cooled, the product was removed from the moulds and evaluated.

20 [0035] The results indicated that in the absence of HPC, the contents of sample A were significantly melted into the candy bleeding color and showing little in the way of particle retention. The samples containing HPC, Samples B & C, retained their shape and color in the candy resulting in a clear candy with discrete particles. When tasted, the candies containing HPC, gave a slight rough surface as the product dissolved, indicating that the extruded flavors has a slightly 25 slower dissolution rate than the other outer candy.

### Example 3

30 [0036] Two versions of a fragrance extrudate were made.

[0037] The first fragrance (A) was made containing 10 weight percent of a fragrance consisting of equal parts of hexyl cinnamic aldehyde, LILLIAL® (International Flavors & Fragrances Inc.), and HELIONAL® (International Flavors & Fragrances Inc.) and was encapsulated in a matrix consisting of maltodextrin, sucrose, maltose and silicone dioxide. Lecithin was included as an emulsifier.

35 [0038] The second fragrance (B) was made containing 12 weight percent of HELIONAL® and was encapsulated consisting of 2% (total product weight) KLUCEL® GF (Hercules Inc.), sucrose maltodextrin cottonseed oil, distilled monoglyceride and silicone dioxide. Lecithin was used as an emulsifier.

[0039] Particles were made and screened to between 30 and 40 US mesh and were dissolved in 66°C water at a level of 0.5 weight percent. The dissolution was monitored via spectrophotometer determination of light transmission. As evidenced by the data in the chart below, the system with the HPC in the matrix released the fragrance at a slower 40 rate.

AQUEOUS % FRAGRANCE RELEASE AT 66°C		
Time (Seconds)	Fragrance A	Fragrance B
0	0	0
10	35	20
20	60	40
30	100	50
40	100	60
60	100	80

55 [0040] This example demonstrates that the system of the present invention is useful in applications that require particles to withstand high moisture and heat during preparation.

Example 4

[0041] In order to evaluate the effectiveness of the present invention, a liquid cherry flavor was added to a hard candy formulation. The candy system was identical except for the form of flavor used. In the control product, liquid flavor was used. In the second candy system, an extruded form of the same flavor was incorporated in which a standard carbohydrate mixture was used as the encapsulating matrix. In the third candy, an extruded form of the same flavor was incorporated in which a carbohydrate mixture containing HPC (KLUCEL®, Hercules Inc.) at a level of 10 weight percent was used as the encapsulating matrix. To each of the candy systems, a liquid flavor content of 0.12 weight percent was added after the candy temperature reached 143°C.

[0042] After processing the flavor, levels remaining in the candy was measured by extraction of the candies followed by GC analysis of the extract. The control candy retained only 2.3 % of the flavoring originally supplied after processing. The candy which had the flavor encapsulated in the standard carbohydrate matrix retained 41% of the original flavor provided to the candy after processing. The candy that had the flavor encapsulated in the HPC retained about 92 % of the flavoring supplied to it.

[0043] The candies were sampled for taste, and the candy which contained the HPC had such a strong cherry flavor that it was determined that the flavor was too strong and that the flavor loading should be reduced. This example demonstrates that the incorporation of HPC in the encapsulating matrix greatly reduces the loss of flavor during processing.

20

**Claims**

1. A composition comprising from 0.1 to 20 weight percent of a flavor or fragrance material encapsulated in matrix comprising from 60 to 99.5 weight percent of materials selected from the group consisting of sugars; maltodextrin having 5 to 20 dextrose equivalent (DE), fats, silicone dioxide, polyhydric alcohols, corn syrup solids, starch, modified starches, emulsifiers and food acids; and from 0.5 to 20 weight percent of hydroxypropyl cellulose.
2. The composition of claim 1 wherein matrix is from 5 to 95 weight percent.
3. The composition of claim 2 wherein the maltodextrin has a dextrose equivalent of from 5 to 15.
4. The composition of claim 3 wherein the maltodextrin has a dextrose equivalent of from 10 to 14.
5. The composition of claim 1 wherein the level of hydroxypropyl cellulose is from 2 to 10 weight percent.
6. The composition of claim 1 wherein the composition has a Tg of greater than 35°C.
7. The composition of claim 1 wherein the matrix comprises from 5 to 75 weight percent starch.
8. The composition of claim 1 wherein the matrix comprises from 1 to 80 weight percent of a food acid.

45

50

55



(19)

Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11)

EP 1 304 044 A3

(12)

## EUROPEAN PATENT APPLICATION

(88) Date of publication A3:  
19.11.2003 Bulletin 2003/47

(51) Int Cl.7: A23L 1/22, A23P 1/08,  
A23P 1/12, A23G 3/00,  
C11D 3/50, A23L 1/00,  
C11D 3/22

(43) Date of publication A2:  
23.04.2003 Bulletin 2003/17

(21) Application number: 02257279.6

(22) Date of filing: 21.10.2002

(84) Designated Contracting States:  
AT BE BG CH CY CZ DE DK EE ES FI FR GB GR  
IE IT LI LU MC NL PT SE SK TR  
Designated Extension States:  
AL LT LV MK RO SI

(72) Inventors:  
• Lou, Wen Chin  
Morris, New Jersey 07950 (US)  
• Popplewell, Lewis Michael  
Monmouth, New Jersey 07751 (US)

(30) Priority: 22.10.2001 US 54239

(74) Representative: Mercer, Christopher Paul et al  
Carpmaels & Ransford  
43, Bloomsbury Square  
London WC1A 2RA (GB)

(71) Applicant: INTERNATIONAL FLAVORS &  
FRAGRANCES INC.  
New York New York 10019 (US)

### (54) Hydroxypropyl cellulose encapsulation material

(57) Hydroxypropyl cellulose is employed to encapsulate flavor or fragrance materials that could be employed in diverse applications such as food products and laundry applications.



European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number  
EP 02 25 7279

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.)
X	EP 1 034 705 A (HAARMANN & REIMER GMBH) 13 September 2000 (2000-09-13)	1-3,5	A23L1/22 A23P1/08
Y	* claims 1,3-6,9; example 1; table 1 * * page 2, paragraphs 7-9 *	6,8	A23P1/12 A23G3/00 C11D3/50
E	WO 02 102173 A (ZERBE HORST G ;AL-KHALIL FADIA (US)) 27 December 2002 (2002-12-27) * claims 1-3,9,16-19; example 1 * * page 3, line 22-26 * * page 4, line 5-14,20 - page 5, line 19 *	1-4,7	A23L1/00 C11D3/22
D,Y	US 6 187 351 B1 (POPPLEWELL LEWIS M ET AL) 13 February 2001 (2001-02-13)	6,8	
A	* claims 1,9,25,26,31; examples 3,5 * * column 1, line 14-18 * * column 3, line 62 - column 4, line 53 * * column 5, line 12-28 * * column 6, line 39-67 * * column 7, line 29 - column 8, line 7 * * column 9, line 34-60 * * column 10, line 29-65 * * column 11, line 36 - column 13, line 14 *	1-5,7	
A	WO 00 59314 A (WRIGLEY W M JUN CO) 12 October 2000 (2000-10-12) * claims 1,4,5,22; examples 1,2 * * page 4, line 1-26 * * page 6, line 18 - page 8, line 11 * * page 8, line 25 - page 9, line 26 * * page 14, line 5-10 *	1,2	A23L A23G C11D
A	EP 0 492 981 A (WM WRIGHLEY JR COMPANY) 1 July 1992 (1992-07-01) * claims 1,2,4-6,29; examples 1,2 * * page 2, line 40-43,51-59 * * page 3, line 8-17,37-41 * * page 4, line 2-19,25-36 *	1,2	
	-/-		
The present search report has been drawn up for all claims			
Place of search:  THE HAGUE	Date of completion of the search  29 September 2003	Examiner  Tallgren, A	
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons S : member of the same patent family, corresponding document	
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document			



European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number  
EP 02 25 7279

DOCUMENTS CONSIDERED TO BE RELEVANT									
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)						
A	WO 94 14330 A (SONG JOO H) 7 July 1994 (1994-07-07) * claims 1-7; examples 1-4 * ---	1,2							
A	WO 95 01166 A (HATA TAKEHISA ;ISHII KYOKO (JP); KADO KAZUTAKE (JP); SAWAI SEIJI () 12 January 1995 (1995-01-12) * claims 1,7; examples 1-5 * * page 1, line 6-10 * * page 2, line 20-11 * ---	1,2							
A	EP 0 277 741 A (TAKEDA CHEMICAL INDUSTRIES LTD) 10 August 1988 (1988-08-10) * claims 1-3,5-7; examples 1,3-11 * * page 2, line 35-50 * * page 3, line 50-56 * * page 4, line 1-4,15-22 * -----	1,2,5,7							
			TECHNICAL FIELDS SEARCHED (Int.Cl.7)						
<p>The present search report has been drawn up for all claims</p> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 33%;">Place of search</td> <td style="width: 33%;">Date of completion of the search</td> <td style="width: 34%;">Examiner</td> </tr> <tr> <td>THE HAGUE</td> <td>29 September 2003</td> <td>Tallgren, A</td> </tr> </table>				Place of search	Date of completion of the search	Examiner	THE HAGUE	29 September 2003	Tallgren, A
Place of search	Date of completion of the search	Examiner							
THE HAGUE	29 September 2003	Tallgren, A							
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ..... & : member of the same patent family, corresponding document							
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document									

**ANNEX TO THE EUROPEAN SEARCH REPORT  
ON EUROPEAN PATENT APPLICATION NO.**

EP 02 25 7279

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report.  
 The members are as contained in the European Patent Office EDP file on  
 The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

29-09-2003

Patent document cited in search report		Publication date		Patent family member(s)	Publication date
EP 1034705	A	13-09-2000	DE	19955131 A1	14-09-2000
			EP	1034705 A2	13-09-2000
			JP	2000297293 A	24-10-2000
WO 02102173	A	27-12-2002	US	2003044511 A1	06-03-2003
			WO	02102173 A2	27-12-2002
			US	2003053962 A1	20-03-2003
US 6187351	B1	13-02-2001	US	5897897 A	27-04-1999
			US	5603971 A	18-02-1997
			US	2003026874 A1	06-02-2003
			US	6416799 B1	09-07-2002
			AU	6701694 A	08-11-1994
			CA	2160684 A1	27-10-1994
			EP	1123660 A2	16-08-2001
			EP	0693884 A1	31-01-1996
			FI	954911 A	16-10-1995
			JP	8509018 T	24-09-1996
			NO	954071 A	13-10-1995
			WO	9423593 A1	27-10-1994
WO 0059314	A	12-10-2000	AU	4025400 A	23-10-2000
			BR	0009488 A	16-04-2002
			CA	2366768 A1	12-10-2000
			CN	1348333 T	08-05-2002
			EP	1173067 A1	23-01-2002
			JP	2002540777 T	03-12-2002
			PL	350925 A1	10-02-2003
			WO	0059314 A1	12-10-2000
			US	2002064576 A1	30-05-2002
EP 0492981	A	01-07-1992	US	5128155 A	07-07-1992
			AT	118309 T	15-03-1995
			AU	640329 B2	19-08-1993
			AU	8977791 A	25-06-1992
			CA	2057781 A1	21-06-1992
			CN	1063602 A	19-08-1992
			DE	69107429 D1	23-03-1995
			DE	69107429 T2	17-08-1995
			DK	492981 T3	10-04-1995
			EP	0492981 A1	01-07-1992
			FI	915837 A	21-06-1992
			JP	2568338 B2	08-01-1997
			JP	5199841 A	10-08-1993
			NO	915033 A	22-06-1992

**ANNEX TO THE EUROPEAN SEARCH REPORT  
ON EUROPEAN PATENT APPLICATION NO.**

EP 02 25 7279

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

29-09-2003

Patent document cited in search report		Publication date		Patent family member(s)	Publication date
WO 9414330	A	07-07-1994	WO	9414330 A1	07-07-1994
WO 9501166	A	12-01-1995	AT	204163 T	15-09-2001
			AU	682192 B2	25-09-1997
			AU	6984294 A	24-01-1995
			CA	2165789 A1	12-01-1995
			CN	1127990 A ,B	31-07-1996
			DE	69427981 D1	20-09-2001
			DE	69427981 T2	29-11-2001
			DK	706380 T3	08-10-2001
			EP	0706380 A1	17-04-1996
			ES	2161768 T3	16-12-2001
			GR	3036407 T3	30-11-2001
			WO	9501166 A1	12-01-1995
			JP	9501150 T	04-02-1997
			PT	706380 T	28-02-2002
			US	5683716 A	04-11-1997
EP 0277741	A	10-08-1988	AT	80292 T	15-09-1992
			AT	150963 T	15-04-1997
			CA	1312548 C	12-01-1993
			DE	3855852 D1	07-05-1997
			DE	3855852 T2	17-07-1997
			DE	3874360 D1	15-10-1992
			DE	3874360 T2	14-01-1993
			EP	0277741 A1	10-08-1988
			EP	0475536 A1	18-03-1992
			ES	2052697 T3	16-07-1994
			ES	2099130 T3	16-05-1997
			GR	3006170 T3	21-06-1993
			GR	3023599 T3	29-08-1997
			HU	46238 A2	28-10-1988
			JP	8032625 B	29-03-1996
			JP	63301816 A	08-12-1988
			JP	2969345 B2	02-11-1999
			JP	11071267 A	16-03-1999
			KR	9605138 B1	22-04-1996
			US	5026560 A	25-06-1991
			US	5516531 A	14-05-1996

**THIS PAGE BLANK (USPTO)**